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PARTITIONING OF SELECTED POLYCHLORINATED BIPHENYLS TO NATURAL SEDIMENTS

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Abstract—The partitioning of two polychlorinated biphenyl mixtures (Aroclor 1016 and 1242) and two tetrachlorobiphenyl isomers (2,5,3',4' and 2,6,2',6' tetrachlorobiphenyl) between water and four natural sediments was evaluated in laboratory investigations. For both Aroclor mixtures, the extent of adsorption was comparable on all four natural sediments studied. For individual isomers, partitioning was comparable with that observed for the mixtures. Both particle size distribution and total organic carbon were important factors in determining the extent of partitioning for the two isomers; but in the case of the mixtures, there was no correlation with organic content.

INTRODUCTION

The occurrence of polychlorinated biphenyls (PCBs) in environmental systems has created concern on the part of environmentalists regarding the fate and transport of these potentially toxic chemicals within natural waters and sediments (Gustafson, 1970; Nisbet & Sarofin, 1972). Existing information on physical properties of PCBs, however, is insufficient to draw satisfactory conclusions regarding their transport within natural systems (Risebrough *et al.*, 1968; Hammond, 1972).

Recent investigations in our laboratory have focused on the physico-chemical characteristics and biodegradation routes of PCBs in relation to the fate and transport of these chemicals in the environment. That sediments and soils are involved in the partitioning and transport of toxic pollutants is well documented; however, neither the transport of PCBs in sediments nor their sorption characteristics is well understood. Haque *et al.* (1974) in their investigation of the water solubility, vapor behavior, and adsorption characteristics of Aroclor 1254 found the mixture to be readily adsorbed onto soil and clay surfaces. Haque & Schmiedding (1976), who investigated the adsorption of selected polychlorinated isomers in somewhat greater detail, showed that these materials were also substantially sorbed to humic acid, clays, and soils and that the degree of sorption increased with the degree of chlorination, i.e. inversely related to the solubility.

The investigation reported here was undertaken to evaluate the relative magnitude of partitioning between water and natural sediments of two commer-

cial polychlorinated biphenyl mixtures (Aroclor 1016 and 1242) containing primarily tri and tetrachlorinated compounds and two chlorinated biphenyl isomers (2,5,3',4' and 2,6,2',6' tetrachlorobiphenyls).

EXPERIMENTAL

Sediment sampling

Bottom sediments from a river and three ponds in north Georgia were collected with an Eckman dredge to a depth of approximately 5 cm. Sediments were transferred to 3.8-l amber glass bottles and kept under refrigeration while transporting to and while in use in the laboratory. Particle size distribution, total organic carbon (TOC), and pH characteristics of the four sediments are shown in Table 1.

Analytical procedures

Aroclor 1016 and 1242 mixtures were provided by Dr. R. G. Webb, U.S. Environmental Protection Agency; the isomers 2,5,3',4' and 2,6,2',6' tetrachlorobiphenyl were obtained from Analabs, Inc., and were 99% pure. The extraction procedure consisted of taking whole samples from isotherm studies, centrifuging them at 10,000 rpm for 20 min, decanting the aqueous phase, and extracting with iso-octane. Quantitative determination of Aroclor 1016 and 1242 in the aqueous phase was made using the procedure of Webb & McCall (1973) and included a comparison of the sum of the major component peaks observed in either 1016 or 1242. Analysis was performed with a MicroTek 220 gas chromatograph equipped with a ^{63}Ni electron capture detector and a 3% SE-30 on chromosorb Q column (2.0 m). The conditions were as follows: I_T , 230°; C_T , 200°; D_T , 300°; flow (N_2), 75–80 $\text{cm}^3 \text{min}^{-1}$.

(Mention of trade names or commercial products does not constitute endorsement or recommendation for use.)

Table 1. Physical-chemical characteristics of sediments

Sediment	TOC (%)	pH	Particle size distribution		
			% Sand	% Silt	% Clay
Oconee River	0.4	6.5	93.0	6.0	2.0
USDA Pond	0.8	6.4	—	—	—
Doe Run Pond	1.4	6.1	56.0	44.0	<1.0
Hickory Hill Pond	2.4	6.3	55.0	45.0	<1.0

Table 2. Partition coefficient of Aroclor's 1016 and 1242 to natural sediments

Source	Partition coefficients $K_p (\times 10^{-3})$	
	1016	1242
Hickory Hill Pond	$1.30 \pm 0.09^*$	$1.25 \pm 0.11^*$
Doe Run Pond	$1.29 \pm 0.08^*$	$1.09 \pm 0.16^*$
USDA Pond	$1.37 \pm 0.14^*$	$1.21 \pm 0.12^*$
Oconee River	$0.62 \pm 0.04^\dagger$	$0.54 \pm 0.04^\dagger$

Values followed by * or † are not significantly different at the 5% level as determined by Duncan's test.

Adsorption isotherms

Aqueous solutions of PCBs were prepared by coating isooctane-PCB solutions on volumetric flasks by evaporating the solvent and equilibrating with sterile, deionized water. Samples were taken prior to isotherm studies to ensure that concentrations of the Aroclor mixtures were at the desired levels. Concentrations of PCBs used in the investigations were maintained at levels below experimentally determined solubilities. In adsorption experiments, the adsorbent and PCB concentrations were varied independently. Adsorption isotherms were determined by equilibrating an aqueous solution of PCB with a known amount of adsorbent material in stainless steel centrifuge tubes and shaking for 2 h. Adsorption equilibrium was achieved quite rapidly (less than 30 min).

Sorbed PCB was determined by difference and the resulting partition coefficient was calculated using the following equation:

$$K = (x/m)/C_{eq} \quad (1)$$

where:

- K = partition coefficient,
- x = mass of PCB in sediment at equilibrium (mg),
- m = mass of sediment (g),
- C_{eq} = mass (mg) of PCB per mass (g) of water at equilibrium.

RESULTS AND DISCUSSION

According to Haque & Schmedding's (1976) observations, partitioning increased inversely with the solubility of the compound. However, the Aroclors used in our study are so similar in their physico-chemical properties that the chromatograms showed no prefer-

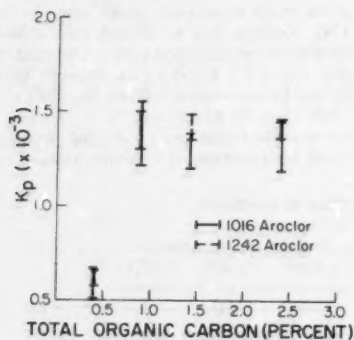


Fig. 1. Range of measured partition coefficients versus organic matter.

Table 3. Partition coefficients for two tetrachlorobiphenyl isomers to natural sediments

Sediment	Partition coefficient $K_p (\times 10^{-3})$	
	2,5,3',4'	2,6,2',6'
Hickory Hill Pond	1.18 ± 0.11	1.27 ± 0.22
Doe Run Pond	0.99 ± 0.08	1.08 ± 0.13
USDA Pond	0.58 ± 0.06	0.65 ± 0.04
Oconee River	0.42 ± 0.09	0.51 ± 0.07

ential sorption of the more highly chlorinated components in the Aroclor mixtures. As shown in Table 2, partition coefficients for the Aroclors were very similar even though Aroclor 1242 is more highly chlorinated, i.e. it contains a higher percentage of tetra- and penta-chlorinated biphenyls. It is possible that experimental errors combined with sediment particle size differences, i.e. bulk bottom sediments versus suspended particulates, may have suppressed our ability to measure small partitioning differences between Aroclors and the bottom sediments.

The three pond sediments had similar partition coefficients with the Aroclor mixtures, but the value for the Oconee River sediment was significantly lower. Based on particle size fractionation results, similarities in partitioning to the three pond sediments is reasonable. The lower partition coefficient for the river sediment is probably indicative of its much higher percentage of sand and coarse silt. Differences in partitioning of the Aroclors related to increasing TOC (total organic carbon) were expected; however, Fig. 1 indicates that partitioning was independent of TOC. The range of TOC within the three sediments was probably too narrow to obtain a clear dependency with the Aroclor mixtures.

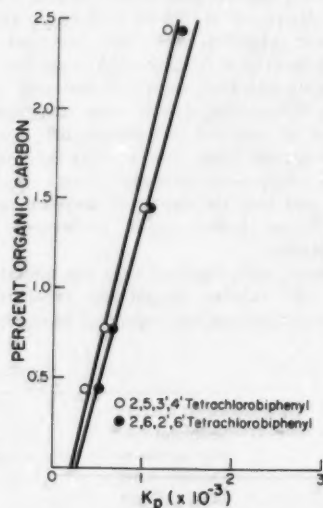


Fig. 2. Measured partition coefficients for tetrachlorobiphenyl isomers versus organic content.

Partition coefficients for the isomers (Table 3) were within the same range as observed for the Aroclor mixtures. Total organic carbon had a more pronounced effect on partitioning than did particle size. Partitioning of the isomers was linear with TOC (Fig. 2). These findings are consistent with reports of increasing adsorption with increasing organic content (Lambert, 1968). The reasons for the difference in behavior between the mixtures and isomers is not known.

From our investigations of the partitioning of PCBs, adsorption and desorption occurred rapidly with the natural sediment. Partitioning of the Aroclor mixtures was independent of sediment type within a factor of 2-3 and independent of TOC. On the other hand, partition coefficients of isomers were linear with TOC and similar to the Aroclors. Both sediment particle size and total organic carbon are important for interpretation of the partitioning behavior of both mixtures and individual compounds. Other PCB mixtures and isomers should partition similarly with sediment.

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